

Stratospheric Albedo Modification by Aerosol Injection

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Abstract

This paper reviews and develops the proposal, widely discussed but not examined in detail, to use stratospheric aerosols to increase the Earth's albedo to Solar radiation in order to control climate change. The potential of this method has been demonstrated by the "natural experiments" of volcanic injection of sulfate aerosols into the stratosphere that led to subsequent observed global cooling. I consider several hygroscopic oxides as possible aerosol materials in addition to oxides of sulfur. Aerosol chemistry, dispersion and transport have been the subject of little study and are not understood, representing a significant scientific risk. Even the optimal altitude of injection and aerosol size distribution are poorly known. Past attention focused on guns and airplanes as means of lofting aerosols or their chemical precursors, but large sounding rockets are cheap, energetically efficient, can be designed to inject aerosols at any required altitude, and involve little technical risk. Sophisticated, mass-optimized "engineered" particles have been proposed as possible aerosols, but the formidable problems of their production in quantity, lofting and dispersion have not been addressed.

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I. INTRODUCTION

Several historic volcanic eruptions (Tambora in 1815, preceding the “Year [1816] without a Summer” in the northeastern US, Krakatau in 1883, El Chicón in 1982 and Pinatubo in 1991) were associated with short-term ($\sim 1\text{--}3$ years) subsequent hemispheric cooling. It has been generally accepted for a long time that the eruptions caused the cooling (and spectacular sunrises and sunsets) by injecting aerosols into the troposphere and lower stratosphere, and that these effects disappeared as the aerosols were removed from the atmosphere by sedimentation or scavenging by hydrometeors.

All aerosols scatter sunlight, reducing the insolation at the surface, and therefore cool the surface and the mixed boundary layer. The scattering, augmenting the Rayleigh scattering of clear air, makes vivid sunsets. Some aerosols (soot and some mineral dusts) also absorb sunlight and heat their surrounding air, and indirectly the ground, but this is believed to be a comparatively minor effect. These properties and effects of aerosols (unlike most of the rest of global change and climatological research) are uncontroversial.

Several decades ago this led to the suggestion¹ that injection of anthropogenic aerosols into the stratosphere could cool the climate, were that desired. The growing concern over global warming, together with the expectation that it will increase with the increase in anthropogenic greenhouse gases, particularly CO₂ but possibly also CH₄ and perhaps others, has led to a revival of interest in the injection of anthropogenic aerosols. A recent extensive review was presented by Rasch, *et al.*², and a critical assessment by Lacis, *et al.*³, who discuss in more detail issues of particle agglomeration and scattering properties that we allude to here.

II. AEROSOL SCATTERING

The most efficient (per unit mass) spherical bulk-density scatterers have radii of about 0.1 of the wavelength of the scattered radiation. For the Solar spectrum, peaking (depending on whether the peak is per unit wavelength or per unit frequency) on the red side of the visible spectrum, this means radii ≈ 1000 Å. Scattering from such particles is described as Mie scattering, and results are widely available. Figure 1 (⁴) shows the scattering efficiency as a function of the particle diameter for a sphere of refractive index $n = 1.42$, characteristic

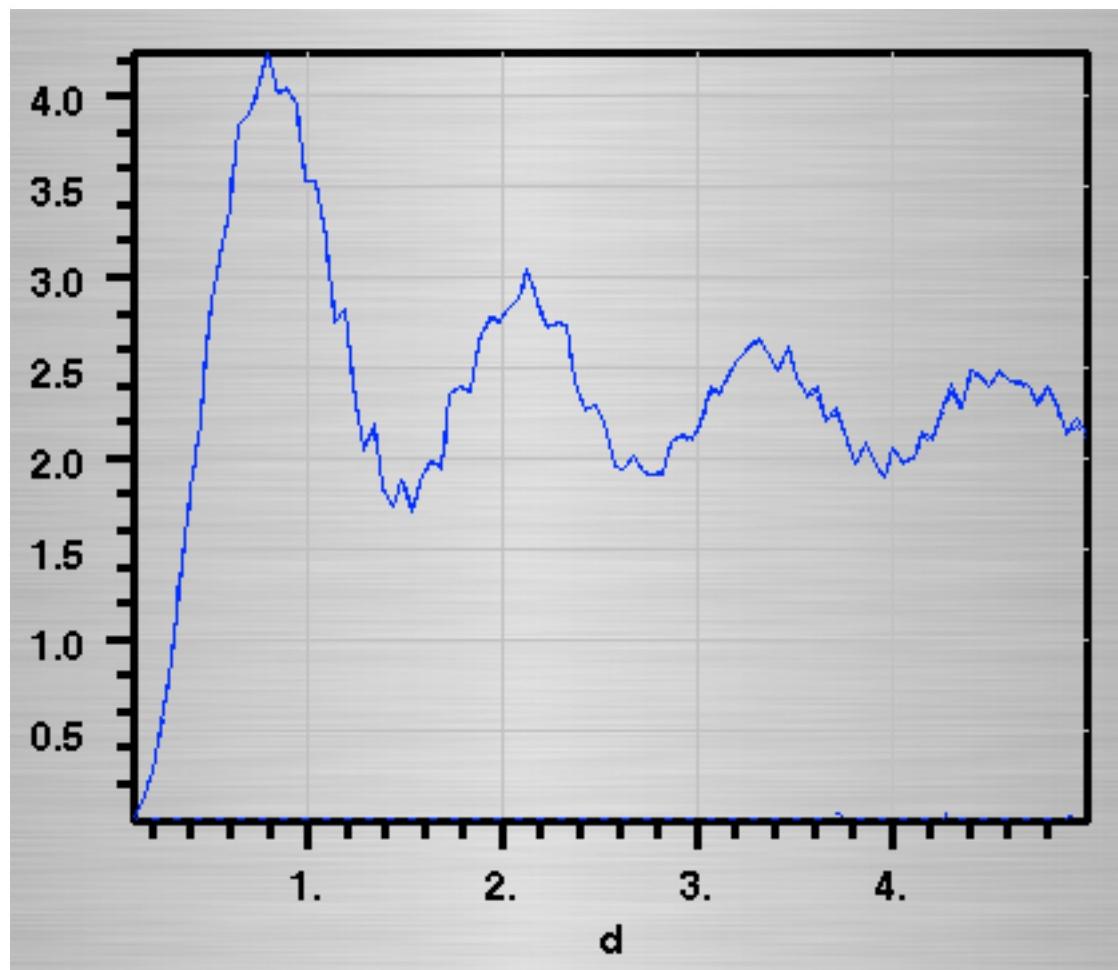
of $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$ ²⁴.

The asymptotic forms of the scattering cross-section are elementary. For $d \ll \lambda$ the cross-section is just the Rayleigh cross-section $\sigma \propto d^6/\lambda^4$, leading to a mass efficiency $\epsilon \equiv \sigma/(\pi\rho d^3/6) \propto d^3/\lambda^4$, so that very small particles are completely ineffective. We have retained the wavelength dependence because it is also strong; particles that are effective in scattering blue light are an order of magnitude less effective in scattering red light (hence the blue sky, or blue diesel exhaust). For $d \gtrsim \lambda$, $\sigma \propto d^2$, (twice the geometric cross-section²⁵), so that $\epsilon \propto d^{-1}$, and large particles are also inefficient users of mass.

III. AEROSOL MATERIALS

Natural scattering aerosols are mostly soot (from fires), minerals (from dust), salt (in the oceanic troposphere, from evaporated spray), water (condensed from the atmosphere) and concentrated sulfuric acid (resulting from oxidation of S to SO_2 to SO_3 followed by hydration). Soot, wind-lofted mineral dust and salt are generally limited to the troposphere, where their lifetimes (against precipitation) are short. Water is in local equilibrium with its vapor (although often not in liquid-ice equilibrium; undercooled liquid drops are very common), and hydrometeors are continually condensing and evaporating, so that any attempt to increase the quantity in the stratosphere would probably be rapidly redistributed through the vapor phase into larger ice crystals that would precipitate. Volcanic cooling is largely the result of mineral and sulfate aerosols lofted into the stratosphere. Their vapor pressures are negligible (in the case of sulfuric acid, this refers to the vapor pressure of the sulfur-containing SO_3) so the larger particles do not grow at the expense of the smaller ones (except very slowly by agglomeration), and lifetimes may be many years, depending on altitude.

For deliberate albedo modification we wish to use aerosols that scatter sunlight with minimal absorption, minimal mass, minimal vapor pressure (so they don't evaporate), are chemically stable in our oxidizing atmosphere, even under the influence of the Solar UV flux at stratospheric altitude, and minimal scattering or absorption of thermal infrared radiation in the 8–14 μ window from the ground (this window is the vent in the atmospheric greenhouse). These criteria point to oxides of elements in the third row of the periodic table (water is excluded by its high vapor pressure) or of boron. Transition metal oxides are



Scattering efficiency (vs. geometric cross-section) as a function of diameter (in units of the wavelength) for a dielectric sphere of refractive index n=1.45 (sulfuric acid).

excluded because they are generally strong absorbers of visible light and second row elements other than boron are excluded because they are either rare and toxic in all chemical forms (beryllium) or greenhouse gases themselves.

Short ($\lambda/2$) fine (thickness and width $\sim 300 \text{ \AA}$) aluminum wires, coated with inert oxides of aluminum or silicon for protection against oxidation, have been proposed⁷ as mass-efficient scatterers, either in the upper atmosphere or in space. Although the mass efficiency is spectacular, these wires must be mounted on much more massive sheets of other materials (perhaps plastic) for handling. In one space application⁷ the wires form a large two dimensional array whose form must be maintained to achieve the desired diffraction pattern, requiring a stiff and massive mount. An area in excess of 1% of the projected surface area of the Earth must be covered with this material, either in space or in the atmosphere. As a rough guide to the thickness of cheap films that can be manipulated, polyethylene (a very cheap plastic of low modulus and low strength) garment bags are about 0.5 mil thick (one manufacturer⁸ quotes thicknesses of 0.43 and 0.65 mil), or more than 10μ . Everyday experience with these bags indicates that much thinner films of plastic are likely to be difficult to handle. Conventional high altitude scientific balloons (zero overpressure) are made of 0.8 mil (20μ) polyethylene. The mass of film required per dipole is likely to far exceed the mass of a dielectric particle of similar scattering cross-section, which is typically ~ 0.01 mil in diameter.

The remaining candidates (some of them are hydrated oxides and some will hydrate further under stratospheric conditions) are

1. Li_2O
2. $\text{B}(\text{OH})_3$
3. Na_2O
4. MgO
5. Al_2O_3
6. SiO_2
7. H_3PO_4

8. H₂SO₄

Sulfuric acid is most often considered because its precursor oxides are the most abundant in volcanic aerosols (a consequence of the fact that, uniquely in this list, they are volatile), and are a source of tropospheric aerosols as a result of burning fuels with sulfur impurities.

A. Choice

We will focus on the oxides of boron, silicon, phosphorus and sulfur (hydrated SO₃ is sulfuric acid). The reason is that from the preceding list these, and only these, have volatile hydrides that are expected to oxidize to the oxides in the stratosphere. Introduction as volatile (gaseous or pressurized or cryogenic liquid) hydrides facilitates their dispersion, minimizes coagulation by delaying oxidation until after they are well-diluted in the stratosphere, and reduces the mass that must be lofted because the oxygen is drawn from the air.

IV. AEROSOL DISPERSION

The stratosphere is a difficult region to reach. Before considering the vehicle delivering the aerosol material (or its precursor) to the stratosphere, we briefly discuss the problem of dispersion, for it would be pointless to deposit a compact mass or large particles of material that would be inefficient scatterers of light or that would immediately fall out of the stratosphere.

It is difficult to grind a solid to the required particle sizes of $\lesssim 0.1 \mu$ so dispersion to these sizes much occur after release. In addition, it is necessary that the particles be dispersed enough that they do not rapidly reaggregate. This is the principal reason why we focus on materials that can be lofted as gaseous precursors that will not agglomerate, and will produce potentially agglomerating particulates only after they have been diluted to low density.

One may consider three possible means of dispersion:

A. High explosives

High explosives may disperse a solid or liquid, but generally not finely enough for our requirements. Although the available shock energy density ($\mathcal{E} \gtrsim 10^{11}$ erg/cm³) exceeds the energy required to disperse individual atoms or molecules, even in a brittle solid nearly all this energy is turned into bulk kinetic energy and shear rates are much too low to fragment it to the required size. A rough argument comparing the energy in the shearing velocity field to surface energies suggests that the characteristic fragment size will be

$$a \sim \left(\frac{\varpi R^2}{\mathcal{E}} \right)^{1/3} \sim 0.01 \text{ cm}, \quad (1)$$

where $R \sim 30$ cm is the overall size of the exploding system and $\varpi \sim 10^2$ dyne/cm is the surface energy (surface tension) of the surfaces that must be formed in dispersion. In addition, the fragments are stopped by the air in a distance $\sim R(\rho/\rho_a)^{1/3} \sim 50R$, where ρ is the material density and ρ_a is the density of the ambient air. This suggests that agglomeration of explosively dispersed solid or liquid particles into sizes too large to be useful would be a serious problem.

B. Combustion

Combustion of solids or liquids, perhaps initially dispersed by high explosives, may produce aerosols. Of the elements considered, Li, Na, Mg, P, Al and S readily burn at atmospheric pressure (Al only if finely divided or strongly heated). An unresolved issue with the production of fine aerosols by combustion of solids or liquids is that near the burning region the particle density is very high and they may coagulate into sizes too large for them to be mass-efficient scatterers.

It is not obvious that these elements would burn at stratospheric densities, two or three orders of magnitude less. Even if they do, agglomeration in the immediate wake of combustion may be a serious problem that would require experimental investigation.

The (very exothermic) burning of boron particles is problematic⁹ even at full atmospheric density because of the tendency of their surfaces to be covered with a tenacious coat of inert oxide. For the same reason it is essentially impossible to burn bulk silicon. In fact, silicon is used in high temperature micro-turbine engines¹⁰ because of its resistance to oxidation (and ease of lithographic fabrication).

C. Slow Oxidation of Gases

Slow oxidation following mixture into the air of gases containing the desired cation in a reduced state may produce the desired oxide aerosols. Because the reaction is comparatively slow, the gas may be widely dispersed and mixed into the air at low concentration before it reacts, avoiding early agglomeration of the particulates. For this reason we restrict attention to the elements boron, silicon and sulfur that have volatile compounds (hydrides).

The use of H_2S as a source of sulfur has been widely proposed. This method may also work with boron (using diborane B_2H_6), phosphorus (using phosphene PH_3) and silicon (using silane SiH_4). The use of these hydrides has the additional advantage (in comparison to making the oxide on the ground and lofting it into the stratosphere) that the oxygen in the oxide and any water of hydration, which represent 77% of the mass of $\text{B}(\text{OH})_3$, 53% of the mass of SiO_2 , 56% of the mass of P_2O_5 and 71% of the mass of $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$, need not be lofted with the cation, but are taken without mass penalty from the surrounding air. $\text{B}(\text{OH})_3$ has the particular advantage that the molecular weight of boron is about 1/3 of that of sulfur or phosphorus and 2/5 of that of silicon.

Hydrides could be lofted as gas (through a chimney, discussed later), as rapidly evaporating cryogenic or pressurized liquids dispensed by aircraft in the lower stratosphere, or as cryogenic or pressurized liquids contained in artillery shells and dispersed (to droplets no smaller than estimated in Eq. 1, and larger if small charges are used) in the stratosphere by explosives. The dispersed liquid drops will evaporate rapidly and mix into the surrounding air, where the vapor will spread by velocity shear and turbulent diffusion.

It is generally accepted that H_2S will oxidize rapidly to SO_2 . At full (sea-level) atmospheric pressure B_2H_6 , PH_3 and SiH_4 ignite or explode readily in air. Hence we are optimistic that oxidation will soon follow evaporation, but the kinetics require detailed investigation, both experimental and theoretical.

All of the candidate gases are quite toxic, but the oxides are all not specifically toxic (although all, except for SiO_2 , are harmful, chiefly because of their acidity, if ingested in quantity). All the gases are major industrial chemicals, routinely handled without harm if proper precautions are taken. They could be manufactured on and lofted from remote uninhabited islands (desirable, in any case, because most of the lofting mechanisms are poor neighbors) at the required latitudes of injection.

D. Choice

For the reasons described, slow oxidation of a hydride precursor (B_2H_6 , SiH_4 , PH_3 or H_2S) is probably the best method of dispersing aerosols. Of these materials, H_2S probably has the optimal combination of lesser toxicity and high ratio of final aerosol mass to lofted precursor mass. Detailed investigation of the oxidation kinetics is required to determine the size distribution of the final aerosols, which is essential to the practicality of any aerosol method of albedo modification.

V. AEROSOL AGGLOMERATION

Agglomeration is the enemy of dispersion. Particles that agglomerate may become too large to be mass-efficient scatterers, and they may become so large that gravitational settling removes them from the stratosphere unacceptably rapidly.

The Brownian mean free path of a spherical particle of density ρ and radius r in air of density ρ_a is

$$mfp_{part} = \frac{4}{3C_D} \frac{\rho}{\rho_a} r \approx 5 \times 10^4 r, \quad (2)$$

where $C_D \sim 1$ is its drag coefficient. This result is valid in Knudsen flow, in which the mean free path of the air molecules $mfp_a \gg r$, as is the case for particles of interest in the stratosphere. When $mfp_{part} \gg r$, as is always valid for such small particles, the coagulation time of uncharged monodisperse particles

$$t_{coag} = \frac{2}{nK}, \quad (3)$$

where n is the particle number density and the coagulation coefficient

$$K = A \sqrt{\frac{24\pi k_B T r}{\rho}} \approx 4 \times 10^{-9} A \text{ cm}^3/\text{sec}, \quad (4)$$

where A is the accommodation (sticking) coefficient and we have taken $\rho = 1.8 \text{ gm/cm}^3$ (sulfuric acid, but not far wrong for any of the materials considered) and $r = 1000 \text{ \AA}$; these results are similar to those of².

A. Early Times

Agglomeration may be a particularly acute problem in the initial stages of dispersion when the particle density is high. This problem is avoided if the cation of the scattering material is introduced as its volatile hydride so that particulates are not formed (because the oxidation kinetics are not instantaneous) until the vapor has dispersed to low density.

B. Late Times

To provide 3% albedo increase with particles with $r = 1000 \text{ \AA}$ and $\sigma \sim 3 \times 10^{-10} \text{ cm}^2$ implies a column density $\sim 10^8/\text{cm}^2$. Distributed over a column 10 km in height, $n \sim 10^2 \text{ cm}^{-3}$, and $t_{coag} \sim 5 \times 10^6/A \text{ sec}$, or $2/A \text{ months}$. This is prohibitively short unless $A \lesssim 0.1$. The observed persistence of volcanic aerosols implies that this is the case, at least for some of their components (not necessarily sulfuric acid).

We have not been able to do a literature search for empirical intrinsic (uncharged) values of A for the materials under consideration. Coatings, either monolayers acquired in the stratosphere or deliberately introduced during production of particles, may keep A sufficiently small.

C. Electrostatic Repulsion

Electrostatic repulsion between like-charged particles is probably more efficaceous. A quantitative evaluation depends on knowing how the charge is distributed on insulating solid particles, and on an energy minimization calculation for conducting particles (such as sulfuric acid) in which charge is redistributed as two particles approach. A reasonable rough estimate for particles each with net charge Q is

$$A \approx \exp\left(-f \frac{Q^2}{2r k_B T}\right), \quad (5)$$

where the charge distribution and mobility factor f is in the range $1/2 < f \leq 1$; $f = 1$ for spherically symmetric immobile charge distributions. Adopting this value, for $r = 1000 \text{ \AA}$ and $A = 0.1$ we find $Q = 1.4 \times 10^{-9} \text{ esu}$, or three elementary charge units. The surface potential is then 0.04 Volt.

Particles are readily charged by friction if they are dispersed from a dense cloud in which they collide frequently, but more than one composition of particle must be present, with different electroaffinities, in order that all or most of the same species have the same sign of charge. This may explain the charging of volcanic particles²⁶, which have a variety of compositions, but will probably not occur for chemically uniform anthropogenic stratospheric aerosols.

In the stratosphere (particularly above much of the ozone layer) particles are likely to be charged by Solar ultraviolet photons, while the electrons attach themselves to molecular species with positive electron affinities. O₂ has an electron affinity EA= 0.44 eV but that of N₂ is negative¹¹. If the kinetics are rapid enough to permit an approach to thermodynamic equilibrium, then the abundant O₂ molecules determine the chemical potential of the electrons. The particles will have a mean positive surface potential equaling the O₂ electron affinity, and $Q \approx EA r$. Then

$$A \approx \exp\left(-f \frac{QEA}{k_B T}\right) \approx \exp\left(f \frac{EA^2 r}{e^2 k_B T}\right) \approx \exp(-538f) \lll 1. \quad (6)$$

Under these conditions there would be no agglomeration. The electrostatic repulsion is a thermodynamic effect, not a kinetic one, so there is no obvious reason why it should be reversed even in the absence of ionizing ultraviolet radiation (nighttime).

The Arrhenius factor for thermal ionization of O₂⁻ of $\exp(-EA/k_B T) \sim \exp(5100^\circ K/T)$ ranges from $\sim 10^{-10}$ at the cold (217°K) tropopause to $\sim 10^{-8}$ at the warmer (271°K) stratopause, so we may expect the electrons to remain bound to O₂ molecules. Neutralization may occur by aerosol-O₂⁻ collisions, at a rate that must be calculated. The lower the density of aerosols, the fewer O₂⁻ and the slower the neutralization, so this process may set an upper bound to the nighttime degree of aerosol ionization. However, nonequilibrium chemical kinetics is complicated and may lead to surprises.

VI. AEROSOL LOFTING

A number of methods of lofting particulates (or, more likely, their chemical precursors, for reasons discussed in the previous section) to stratospheric altitudes may be considered. The feasibility of each depends on the material being lofted and on the means of dispersion, so these three lists are not independent.

A. Aircraft

Injection of megaton quantities of sulfuric acid precursors into the stratosphere requires heavy lift aircraft that can fly at these altitudes. The KC135 has a service ceiling of 50,000' (17 km), at the lower edge of the equatorial stratosphere. The USAF has an inventory of several hundred, 161 of which (along with 38 similarly capable B52s) are scheduled for retirement in the next few years. Gaskill¹² has suggested their use to introduce aerosols or their precursors into the stratosphere. Other heavy lift aircraft, such as commercial airliners, are not capable of reaching these altitudes.

The chief limitation of the KC135 and B52 is their altitude. It may be that to obtain sufficiently long aerosol residence times altitudes of 30–50 km are required. These altitudes are essentially unreachable by aircraft. The presence of equatorial stratospheric upwelling suggests that these higher altitudes may not be necessary, but only a detailed transport calculation, founded on empirical velocity field and turbulent transport data, combined with tracer experiments, can answer this question.

B. Guns

Naval guns have been proposed as a means of stratospheric lofting. The summit of the naval artillery art was achieved in the Iowa class battleships, whose $50 \times 16''$ guns²⁷ fired a 2700 lb shell with a muzzle velocity of 2500 ft/sec (762 m/sec) at a firing rate of two shells per minute¹³. In the absence of an atmosphere, such a shell fired upwards would reach an altitude of 29.6 km.

Air drag reduces the muzzle velocity by a multiplicative factor

$$\frac{v_f}{v_i} = \exp \left[-\frac{C_D}{2} \left(\frac{1000 \text{ gm/cm}^2}{BC} \right) \right] \approx 0.90, \quad (7)$$

where we have taken a mean drag coefficient $C_D = 0.2$, used the ballistic coefficient of the shell $BC = 944 \text{ gm/cm}^2$ and made the approximation that the atmospheric scale height is small compared to the altitude reached. Drag then reduces the attainable altitude to 24.0 km. This may be adequate for injection into the upwelling lower equatorial stratosphere, but would lead to short residence times in the downwelling polar stratosphere.

The main gun on the M1 tank has a muzzle velocity up to 1.7 km/sec, illustrating what can be achieved with conventional chemical propulsion. This might naively suggest a

maximum altitude of 92 km (taking the same value of BC). However, even when scaled to 16" (406 mm) caliber from 120 mm, this high muzzle velocity is only achieved with a much lighter projectile and a ballistic coefficient less by a factor of 3–7, depending on which of the variants of the M829 ammunition is used for comparison¹⁴. For example, the 10 kg M829A3 round has a muzzle velocity of 1.555 km/sec. Scaled to 406 mm, it would have $BC = 299$ gm/cm². Because air drag would be more important, it could reach an altitude of 63 km (rather than 92 km) with a total shell mass of 387 kg, of which 300 kg might be payload. Optimal design of a gun for stratospheric injection to 30–50 km requires engineering tradeoffs among these factors, but these altitudes are clearly achievable.

With the demonstrated firing rate of two rounds per minute for 16" naval guns, a nominal 1 MT/yr injection rate would require three guns firing at their maximum rate. Tank guns have a firing rate roughly ten times higher, but it is unclear how this would scale to 16" caliber, at which the scaled tank round would carry 1/3 the payload of the naval gun. The optimal gun would lie somewhere between these limits, depending on the desired altitude of injection.

The barrels of tank guns must be replaced every few hundred rounds because of erosion, but this is likely to be less frequent at the lower muzzle velocity we require. The chief consequence of barrel erosion is reduced accuracy, which is not an issue for geoengineering, so it is probably safe to assume that each barrel is capable of firing 1000 rounds, and possibly many more. A nominal 1 MT/yr system with 300 kg payloads per round would fire about 3,000,000 rounds/yr from a total of three guns, and would consume no more than 3000 barrels/yr, corresponding to three barrel replacements daily per gun, and perhaps many fewer.

The cost of the shells (not including the payload) may be $\sim \mathcal{O}(\$10,000)$. This is only a guess as to the cost of these very simple shells, and is meant to be conservative. For comparison, the technically sophisticated JDAM guidance package is estimated to cost \$20,000 per item¹⁵. Mass production would reduce the unit cost far below that typical of low production-run peacetime military systems. This nominal cost corresponds to a lofting cost of \$30/kg, or \$30 billion/MT. More massive shells with parameters closer to those of the extant 16" naval guns would probably have a lower cost per unit mass lofted. The guns, barrels, and other components are likely to be a small fraction of the cost of the shells because of the economies of using them continually over a long period. Unlike military systems, no elaborate turret

capable of aiming accurately over wide angles would be required; the geoengineering guns simply point up.

1. *Davis guns*

Davis guns are (in principle) recoilless; if their barrel is rifled they are also known as recoilless rifles. The absence of recoil is achieved with a barrel open at both ends, with a wad of soft material expelled out the back to take up the recoil of the projectile. They were first developed by CMDR Davis, U. S. Navy, in 1912–14 in order to solve the problem of mounting a large-caliber gun on a fragile airplane. A modern 12" Davis gun operated for many years at the Tonopah test range, firing downward to study earth penetration. Because of their small recoil, Davis guns are capable of firing massive projectiles without enormously robust and expensive mounts, but because of the recoil mass they are energetically inefficient and unlikely to be able to loft material into the stratosphere.

C. Rockets

The required velocities 1–1.5 km/sec are a fraction of the exhaust velocity v_e (usually parametrized as specific impulse $\text{ISP} \equiv v_e/g \approx 300$ s) of solid rocket fuels. The payload mass fraction $1 - \exp -v_b/v_e \approx 0.6\text{--}0.7$, where v_b is the rocket velocity at fuel burnout (exhaustion), of the launch weight. This is greater than for high velocity tank rounds, in which the propellant mass exceeds the projectile mass, but probably less than that of naval gun rounds.

Air drag is less important for rockets than for guns because rockets may be slenderer than gun-launched projectiles, increasing their BC , and because the burnout velocity of a rocket is only achieved above the denser parts of the atmosphere. Additional advantages of rockets include a milder launch environment, permitting the payload to be carried in a thin-walled vessel rather than a massive artillery shell, and the absence of a massive breach and barrel to contain the confined burning propellant. We cannot make a quantitative estimate, but expect the cost of rocket lofting to be substantially less than that of guns²⁸.

D. Balloon-Supported Chimneys

It may be possible to inject gases into the stratosphere through a tube suspended from a balloon²⁹. Such a tube resembles a very high chimney, but must be suspended from a balloon at its upper end. The buoyancy of the balloon must be sufficient to support the weight of the tube. It must also prevent tropospheric winds from turning the tube horizontal, thereby pulling its upper end below the required injection altitude. This section contains some very rough estimates, and is no substitute for an engineering design study.

1. *Chimney materials*

The materials considered have been high molecular weight polymers such as Spectra, liquid crystal polymers such as Vectran, and aramids such as Kevlar. These materials all have uniaxial (aligned fiber) strength ≈ 30 KBar, Young's modulus ~ 1 Mbar and density 1–1.5 gm/cm³¹⁶. If the fibers are distributed orthogonally in a thin sheet its biaxial strength and modulus along the axes may approach half the uniaxial value, but it may be very weak in diagonal tension in which fibers can slide over one another. If the fibers are isotropically distributed very few will be aligned in any direction and the material's tensile strength will depend on their resistance to sliding, not on their uniaxial tensile strength. The loads on the tube will be predominantly along its length, so its fibers may be oriented in that manner. However, it is unclear what tensile strength to assume for a balloon fabric in isotropic plane tension. We parametrize the strength by $S \equiv Strength/(10\text{ Kbar})$. For the tube $S \sim 1$ may be reasonable, but for the balloon S may be much smaller.

2. *Flow through the tube*

Using standard methods¹⁹ we have made a rough estimate of the tube diameter required to accommodate the nominal 1 MT/yr (3×10^4 gm/sec) flow through the tube, assuming a driving pressure roughly comparable to ambient, as will be driven by buoyancy if the molecular weight of the gas is a fraction of that of air. This condition is satisfied if the hydride gas is diluted two- or three-fold with hydrogen. The result is a radius at the upper end of the tube, assuming a pressure there of 30 mbar (about 30 km altitude), to deliver 3×10^4 gm/sec (5×10^7 m³/day) of $r_t \approx 2$ m. The lower end may be several times narrower

because of the higher density there.

3. Aerodynamic loads

The tube passes through the troposphere into the stratosphere, and will occasionally encounter the jet stream. The aerodynamic load for a length L immersed in a jet stream of speed $v = 50$ m/sec is

$$F = C_D L v^2 r_t \rho_a \sim 5 \times 10^{11} \text{ dyne}, \quad (8)$$

where we have taken $L = 1$ km as the jet stream depth, $C_D = 1$ and $\rho_a = 1 \times 10^{-3}$ gm/cm³ for the upper troposphere. It may be possible to reduce this load by a factor 2–3 if the tube is aerodynamically shaped with a “weather vane” to turn it into the wind. In addition, we have made the very conservative assumption that the tube has a constant diameter. In fact, the portion at jet stream altitudes may be a few times narrower because of the higher density there (we have tacitly assumed the gas in the tube to be in pressure equilibrium with the ambient air), reducing the aerodynamic load in proportion.

The cross-section required to bear this load in tension is $50 C_D / S$ cm². The weight of a tube of length $L_t = 50$ km is then

$$W = gM = \frac{g\rho_t L_t C_D}{S} 4 \times 10^{11} \text{ dynes}. \quad (9)$$

Of course, the previous calculation is not self-consistent. The tube must bear its own weight as well as that of any aerodynamic load. We could solve the self-consistent equation, but instead make the following qualitative points:

1. The balloon must support a load $\sim 10^{12}$ dyne.
2. It is essential that the along-axis tensile strength of the tube material be $\mathcal{O}(10)$ Kbar.
3. Minimizing C_D by aerodynamic shaping and optimal orientation of the tube has large benefits.

We conclude that the chimney may be feasible, but involves significant technical risks in material and aerodynamic performance.

E. Photophoresis

Photophoresis (lift provided by asymmetric surface properties of a small oriented particle not in thermal equilibrium with its gaseous environment) has been suggested as an explanation of the presence of tropospheric soot in the stratosphere^{20,21}. Keith²² has suggested its application to deliberately engineered aerosols for the purpose of geoengineering. Photophoresis requires a significant temperature difference between the particle and the gas, particles of low density, and particles with an asymmetric thermal accommodation coefficient and an offset between their center of mass and center of drag.

Pueschel, *et al.*²¹ found that “fluffy” soot aggregates with mean densities of a few tenths of a gm/cm³ could, if sufficiently asymmetric, have sufficient diurnally averaged photophoretic lift to overcome gravity. However, the particles required to increase the Earth’s albedo must not (unlike soot) absorb a significant amount of Solar radiation, and are expected to have higher densities (1.8 gm/cm³ for sulfuric acid, and somewhat greater for other materials). Sulfuric acid aerosols would be spherical liquid drops without any surface asymmetry or offset between their centers of gravity and of drag. Net photophoretic lift appears unlikely for aerosols produced by the processes of Section IV.

Carefully engineered particles²² might do much better. However, their scattering properties are not likely to be a great improvement over those of mineral or liquid particles of similar dimensions, so for them to be useful it must be possible to fabricate and disperse them in megaton quantities at reasonable cost.

F. Choice of Lofting Mechanism

Two of the lofting concepts considered, rockets and guns, are technically mature and would only require engineering development. Rockets may be substantially cheaper. Chimneys would require extensive research and development, and it is difficult to estimate their cost. Photophoresis raises major questions of the ability to engineer and mass-produce suitable particles that do not absorb visible light but have sufficient photophoretic lift to loft them into the stratosphere.

VII. BALLOONS

Balloons have at least three potential applications in geoengineering.

1. A means of carrying material to stratospheric altitude. The material might be a gas (such as a hydride precursor of aerosols) filling the balloon itself, or in a vessel hanging from the balloon.
2. A source of lift for a chimney, as discussed in VID.
3. As reflective objects that themselves modify the Earth's albedo..

These three applications require different kinds of balloons meeting different technical criteria. They are best considered according to their required lifetimes rather than according to their application.

A. Short-lived balloons

The buoyancy of a balloon in pressure equilibrium with the ambient air is

$$B = g(M_a - M_b) = gM_a \left(a - \frac{\mu_b}{\mu_a} \frac{T_a}{T_b} \right), \quad (10)$$

where M_a , μ_a and T_a are the mass, molecular weight and temperature of the displaced air and M_b , μ_b and T_b are the mass, molecular weight and temperature of the gas filling the balloon. Because if $T_a = T_b$ B is independent of altitude (equivalently, independent of atmospheric pressure) such a balloon has no equilibrium height. If B exceeds the load it will rise indefinitely, until (if open at the bottom) it spills lifting gas, or (if closed) it bursts from internal overpressure once the skin expands to its maximum volume. If B is less than the load it sinks to the surface of the Earth.

In practice, the altitude of a pressure-equilibrium balloon, such as those used to loft scientific payloads to the stratosphere, is controlled by dumping ballast. If the filling gas were always in thermal equilibrium with the air it would remain at a constant altitude indefinitely, once enough ballast had been dumped (or gas spilled) that the lift equals the load. But because of the diurnal variation in Solar heating of the skin (and advective heat transport to the filling gas) T_a/T_b varies and ballast or gas must be expended daily. As a result, pressure-equilibrium balloons have flight durations of $\mathcal{O}(10)$ days, except during polar

summer and winter (“midnight Sun” or “noontime night”) when there is no diurnal Solar heating cycle. Somewhat longer durations may be obtained if they are made of material that is less absorbing of Solar near-infrared radiation than polyethylene, in order to reduce the magnitude of their diurnal temperature swings, if they are baffled inside to reduce transport of heat from the skin, or if they are aluminized to reflect sunlight.

These balloons must be in pressure equilibrium with the ambient air because they are made of very weak material (typically 0.8 mil polyethylene, with a tensile strength of \sim 300 bars and even lower yield threshold, so that a 100 m radius balloon begins plastic flow at an overpressure of \lesssim 100 dyne/cm², about 10^{-4} bar or about 10^{-2} of a float pressure of 10 mbar at about 120000'). Such a balloon is very cheap and light, and they have been used to loft scientific payloads for many years.

1. Delivery vehicles

Short-lived balloons are satisfactory if the goal is only to deliver materials to the stratosphere. Gaseous material may either be mixed with hydrogen or helium as the lifting gas, or (if liquid or solid) may be suspended from the balloon, as are scientific payloads. Volatile materials (such as the precursor hydrides we have considered) are better carried as gases to take advantage of their buoyancy which, at least partially, offsets their weight. This also avoids the need to lift the parasitic weight of a cryogenic or pressurized container.

Balloon delivery of materials has been considered and rejected on the grounds that the number of balloons is excessive and that the large number of expended balloons falling to the Earth would pose an unacceptable risk to the environment². Balloons that failed to vent or burst in the planned location might also pose a risk to aviation upon their unpredictable descent. These objections are difficult to evaluate.

A typical scientific balloon operation may cost several hundred thousand dollars, and lofts a payload of order a ton, suggesting a cost per unit mass perhaps 1–10 times that of artillery lofting. The launch of such a balloon is a tricky operation that depends on favorable weather (low wind) at the launch site.

B. Long-lived balloons

If we wish to use a balloon to support a chimney, or to effect a long-term reduction in the Earth's albedo, we must avoid daily expenditure of lift gas or ballast. The solution to this problem is an overpressure balloon, whose volume is essentially independent of its temperature. The concept is old, but its realization has depended on the development of better materials^{17,18}.

1. Lift

To provide 10^{12} dynes of lift at the 30 mbar level requires a volume of about 2×10^{13} cm³, or a radius $r_b \approx 170$ m. The overpressure ΔP it must support is a fraction f_{var} (the fractional diurnal temperature variation) of ambient, or $\sim 10^4$ dyne/cm². The required wall thickness is

$$\Delta r_b \geq \frac{f_{var}P}{2S}r_b \approx \frac{0.01}{S} \text{ cm.} \quad (11)$$

The ratio of the weight W_{skin} of its skin to its buoyant lift is then

$$\frac{W_{skin}}{B} = \frac{3}{2} \frac{f_{var}P}{10^{10}S} \frac{\rho_{skin}}{\rho_a(1 - \mu_b/\mu_a)} \approx 0.05/S, \quad (12)$$

where we have taken a temperature of 250°K, $\rho_{skin} = 1.5$ gm/cm³, $f_{var} = 0.3$ and H₂ as the filling gas.

The importance of the material strength is evident. For example, Mylar has an ultimate tensile strength of 1.5 Kbar²³, which gives $W_{skin}/B \approx 0.3$, so the requirement to contain the overpressure of large temperature swings would exact a large price in a Mylar balloon's lifting capability. The materials discussed in VID are much stronger, but their behavior when used to make membranes subject to isotropic tension must be understood.

2. Balloon Albedo

It is also possible to consider using balloons themselves to increase the albedo of the Earth if they are coated with a material with high reflectivity⁷. The obvious choice is vapor-deposited aluminum. Aluminized plastic films are widely used in applications ranging from insulation (where the aluminum layer inhibits radiative transport of heat) to space flight.

A layer of ~ 300 Å of aluminum is sufficient to reflect most incident Solar visible and near-infrared radiation, while transmitting most of the upwelling mid-infrared radiation of the Earth.

The minimum size of such a balloon is set by the requirement that the aluminum, which contributes negligibly to its strength, have a weight small compared to that of the underlying plastic. This implies $\Delta r \gtrsim 0.3\mu$. The thinnest plastic films of which we are aware are 0.9μ thick. The films used in Solar sail experiments have been either 5μ Mylar or 7.5μ Kapton. We do not know how thin films can be made from the high-strength materials discussed in Section VID. For $\Delta P = 10^4$ dyne/cm², $r = 2 \times 10^6 S = \frac{\Delta r}{1\mu} S \times 200$ cm, assuming the balloon is designed to minimize W_{skin}/B . Smaller balloons are possible (letting S be the maximum achieved tensile stress in the skin, rather than the material's limiting stress), but the minimum radius for which the buoyancy is positive is about 5–10 cm.

Unless there is a breakthrough in making and handling ultra-thin films, the minimum diameter of an overpressure balloon will be ~ 10 –20 cm. Its mass would be ~ 0.5 gm. Although very light, it would be large and very strong. A rain of such balloons that have reached the end of their lives would be a significant hazard to wildlife and conceivably to aviation.

VIII. RESEARCH PROGRAM

This report has discussed a number of questions involving aerosol properties (entirely apart from our understanding of climate, either natural or subject to anthropogenic forcing) that must be answered before it can be known if aerosol mitigation of the thermal effects of increasing atmospheric CO₂ is feasible. We list a number of issues in basic science, each of which needs both theoretical and experimental investigation:

1. Chemical kinetics of oxidation of gaseous hydride precursors
2. Physical kinetics of aerosol aggregation
3. Aerodynamics and aeroelasticity of chimneys and balloons
4. Properties of candidate chimney and balloon materials
5. Stratospheric transport aerosols

- (a) Wind fields
 - (b) Turbulent diffusion
 - (c) Photophoresis
 - (d) Sedimentation
6. Engineered aerosols
7. Side effects of anthropogenic stratospheric aerosols
- (a) Stratospheric chemistry (ozone depletion?, *etc.*)
 - (b) Ecological consequences of increased diffuse (scattered) radiation flux
 - (c) Tropospheric and terrestrial effects of precipitated aerosols

In addition, there are many engineering design issues that must be addressed before any aerosol climate modification plan can be developed. We believe the basic science questions should be answered first, so that the engineering efforts can be directed in a most productive manner.

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- ²⁵ For macroscopic objects half the scattering is into forward angles $\mathcal{O}(\lambda/(\pi d))$ and is not readily observed, so we are familiar with the geometric cross-section only.
- ²⁶ As evidenced by the lightning that accompanies volcanic eruptions.
- ²⁷ The first number is the barrel length in units of the caliber.
- ²⁸ Given these arguments, why have militaries mostly used guns, except at very long ranges? The reasons are: 1. Rockets require sophisticated technology to guide them to a target, while guns are simply pointed (allowing for gravity, air drag and wind). 2. A gun-launched projectile has its full velocity out of the muzzle, while a rocket may not have a lethal impact, and may even be so slow as to be avoidable by the target, until a substantial distance from its launch. None of these arguments apply to geoengineering
- ²⁹ A tube would be required because if the energy injected in the tropopause's Brunt-Visälä time (~ 30 sec) is less than tens of megatons, any unconfined tropospheric injection would mix in the troposphere and not reach the stratosphere. Volcanoes are sufficiently energetic to avoid this, but continual injection is not.